STUDIES ON HOT-MELT PREPREGGING OF PMR-II-50 POLYIMIDE RESIN WITH GRAPHITE FIBERS

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ABSTRACT

A Second generation PMR (*in situ* Polymerization of Monomer Reactants) polyimide resin, PMR-II-50, has been considered for high temperature and high stiffness space propulsion composites applications for its improved high temperature performance. As part of composite processing optimization, two commercial prepregging methods: solution vs. hot-melt processes were investigated with M40J fabrics from Toray. In a previous study a systematic chemical, physical, thermal and mechanical characterization of these composites indicated that poor resin-fiber interfacial wetting, especially for the hot-melt process, resulted in poor composite quality. In order to improve the interfacial wetting, optimization of the resin viscosity and process variables were attempted in a commercial hot-melt prepregging line. In addition to presenting the results from the prepreg quality optimization trials, the combined effects of the prepregging method and two different composite cure methods, i.e., hot press vs. autoclave on composite quality and properties are discussed.

KEY WORDS: Carbon Fiber, Polyimides, Prepreg

1. INTRODUCTION

High temperature polymer composites are required to increase thrust to weight ratios for many propulsion applications. Graphite fiber polyimide composites are well suited for these applications. A collaborative project between NASA Glenn Research Center and Boeing Rocketdyne focuses on propulsion components that are candidates for *Access to Space* applications (1-4). A Second generation PMR polyimide resin, PMR-II-50 (5-9), was considered for these high temperature and high stiffness space propulsion applications, especially in face-sheet sandwich structures.

Resin and solvent content in prepreg are important factors for producing high quality composite components (3-4, 10-13). However, polyimide composites are typically difficult to process due to high viscosities, volatile by-products, a limited availability of high temperature processing materials and expensive tooling. In order to determine if there is a correlation between composite quality and the method of prepregging, this study examines two prepregging methods: solution and hot-melt. Both methods are currently available for PMR resin systems. Additionally, each process uses solvent such as methanol to dissolve the monomer reactants and provide the appropriate prepolymer viscosity to coat the graphite fiber. The solution method, however, uses a more dilute solution (40-60 monomer wt %) than hot-melt prepregging (70-95 monomer wt %).

There are advantages and disadvantages for each prepregging method, even though PMR prepregs have been conventionally made from the solution method. Moreover, processing techniques, using either autoclave or compression molding, are well established for PMR prepreg. Hot-melt prepreg contains significantly less residual solvent than solution prepreg and is somewhat safer to handle. However, if the same well-established processing parameters used for producing PMR composites from solution prepreg are employed for hot-melt prepregs; the reduced solvent content in hot-melt prepregs reduces prepolymer flow, fiber wetting and composite quality. In addition, the principle mechanisms and findings from this study, especially the effects of resin viscosity and wetting behavior on composite quality and properties can be directly applied to other advanced composite manufacturing processes, such as Resin Transfer Molding (RTM) or Resin Film infusion (RFI) that also utilize high concentration solution or resin system (14-15).

Previous studies (3-4) compared the two prepregging processes using PMR-II-50 and three different carbon fiber types. From these studies the following results were found:

- Prepreg from the hot-melt process had significantly less solvent and was "boardy" or dry. HPLC analyses showed that the resin had more aging products such as mono and bis amides. Yet, the time-temperature-viscosity profiles of prepreg that mimicked composite cure cycles were similar between the two prepregs
- In general, physical, structural and mechanical characteristics of composites made with hot-melt prepreg were inferior regardless of curing optimization, e.g., higher void content, lower interlaminar shear strength, and lower OHC modulus and strength
- The critical controlling mechanism identified for the composite quality was resin-fiber interfacial wetting, that is primarily controlled by solution viscosity
- Poor wetting resulted in lower composite mechanical properties and poor thermostability
- The enhanced interfacial wetting was confirmed as the primary reason why composites made of solvent re-saturated hot-melt prepregs showed good quality, good mechanical properties and better thermal stability compared to the solution-prepreg composites. This also suggested that the small quantities of aging products in hot-melt prepreg didn't play a major role in determining composite quality.

It should be emphasized that the prepregging process of PMR type polyimide resins is a crucial step in the production of high quality composites in that the wetting of the reinforcement is determined during prepregging. In essence, there is a balance between the solvent content, fiber wetting, and void formation; wherein solvent is needed for fiber wetting. Yet, too much residual

solvent will induce void formation. Finding this balance for both hot-melt and solution prepreg is the focus of this paper.

2. EXPERIMENTAL

2.1 Design of Experiment Figure 1 illustrates the overall program strategy. The process variables studied for the prepregging optimization of M40J/PMR-II-50 fabric composites were:

- Prepregging method: solution vs. hot-melt
- Monomer solid content (MSC) in resin solution; 45, 79, 82, and 91%. From the MSC-viscosity plot reported earlier (3-4), the solution viscosities of those resins at 32.5 °C were roughly 15cps, 1500cps, 3000cps, and ~9000cps, respectively
- Hot-melt process repeat run number: single pass vs. double pass, i.e., either the prepregging was completed in one pass or repeated twice on the same process line
- Composite processing: hot-press vs. autoclave molding that the consolidation pressure difference was 3.45 MPa (500 psi) and 1.38 MPa (200psi).

The test matrix is summarized in Table I and each combination is identified by a processing code for the purpose of convenience throughout this paper. Composites were evaluated by methods that are sensitive to fiber-matrix interfacial interactions and should be indicative of fiber wetting as listed in Figure 1.

2.2 Materials The PMR-II-50 resin solutions were prepared by Maverick Corp., in Blue Ash, OH. The procedure was noted previously (4). The monomer solid concentrations (MSC in wt percent) of the starting resin solutions were prepared at 50-60 % for the solution prepregging and about 70% for hot-melt prepregging. During prepregging, resin formulators often use an imidized solid content (ISC) to indicate the solution concentration and calculate the prepreg resin content based on fully imidized solids or ISC. The unit of measure is different from MSC in which the volatile loss due to imidization is not included. This difference can be significant since a considerable portion of monomer solids is lost as volatile by-products, 16.6% or 7.8% depending on reaction type, in addition to the total loss of solvent during imidization reaction. The carbon fiber selected for this study was M40JB with density of 1.77 g/cm^3, Young's modulus of 54.7Msi, and strain-to-failure of 1.2%. Fibers in 6k-tow were purchased from Toray Carbon Fibers America, Inc., Santa Ana, CA. The fibers were woven into a 4HS fabric form with a fiber areal weight (FAW) of 215 gm/m^2 at Sigmatex High Technology Fabrics, Inc., Benicia, CA. M40J had an epoxy sizing and was used as received.

2.3 Prepregging Processes Solution prepreg was produced at NASA Glenn. The resin solution was first diluted to about 45 to 50 % MSC by adding methanol and then applied to a precut fabric section (61 cm × 61 cm) with a brush. Brushing was done carefully for even distribution of resin, not to disturb fabric alignment or to introduce any damage on fabric. This technique was compared with commercial prepreg produced at J.D. Lincoln where they passed the fabric through a resin solution bath as described in a previous study (3-4). In order to gradually reduce the solvent content of this prepreg, swatches were dried at room temperature in a hood overnight. This prepreg can be stored in a freezer for up to six months before processing into composites.

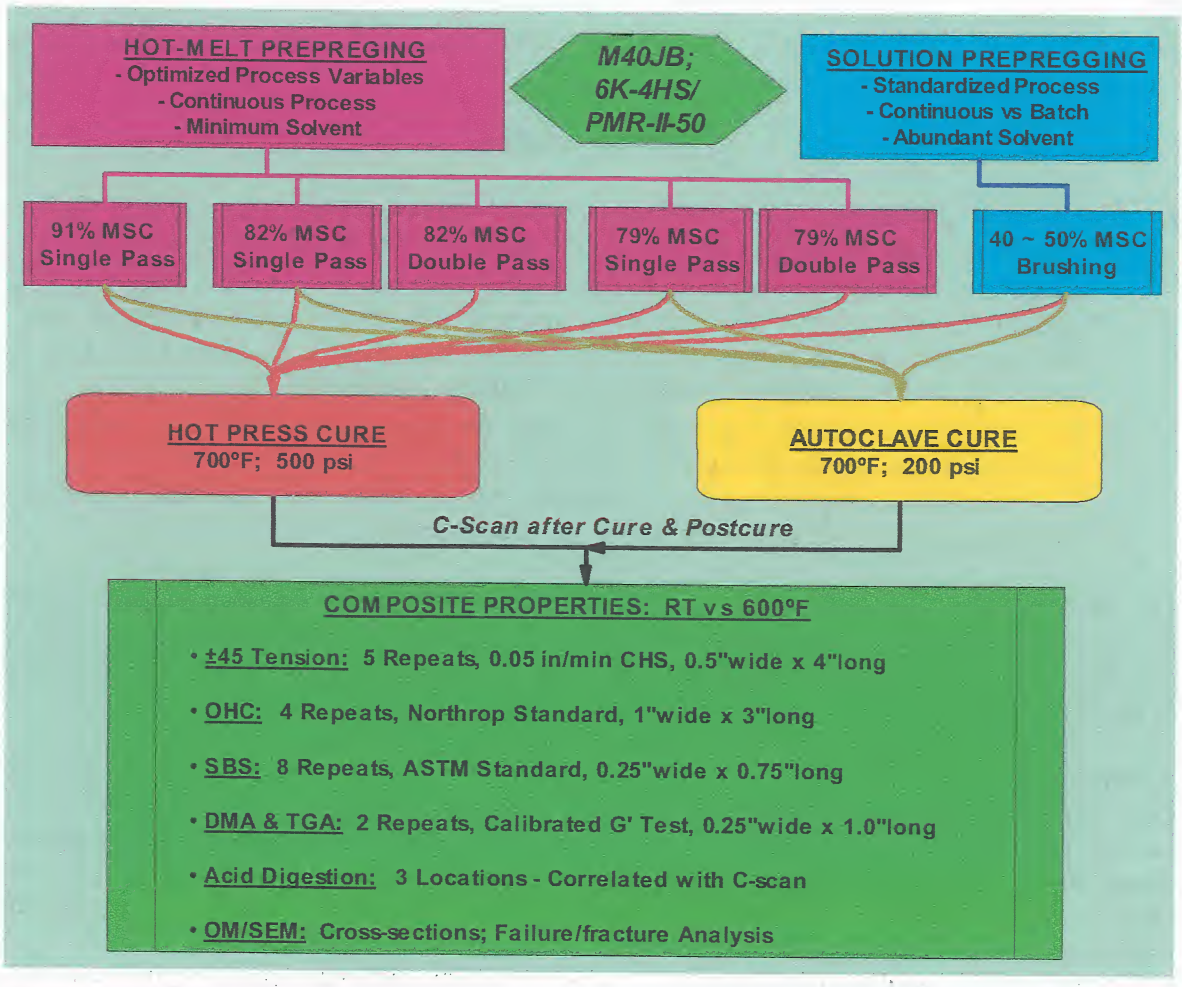

Figure 1 Overall Program Plan

Table I Overall Test Matrix and Processing Code Identification

Processing Code	Prepregging Process		Composite Process
	Method	Resin Solid Content	
SP-P	Solution Prep.	45-50% MSC	Hot-press @ 500psi
SP-A	Solution Prep.	45-50% MSC	Autoclave @ 200psi
HP91S-P	Hot-Melt: Single Pass	91% MSC	Hot-press @ 500psi
HP91S-A	Hot-Melt: Single Pass	91% MSC	Autoclave @ 200psi
HP82S-P	Hot-Melt: Single Pass	82% MSC	Hot-press @ 500psi
HP82D-P	Hot-Melt: Double Passes	82% MSC	Hot-press @ 500psi
HP82S-A	Hot-Melt: Single Pass	82% MSC	Autoclave @ 200psi
HP79S-P	Hot-Melt: Single Pass	79% MSC	Hot-press @ 500psi
HP79D-P	Hot-Melt: Double Passes	79% MSC	Hot-press @ 500psi
HP79S-A	Hot-Melt: Single Pass	79% MSC	Autoclave @ 200psi

The hot-melt prepregging was conducted at YLA Incorporated in Benicia, CA using two different lines, a 60″ wide commercial (Figure 2) and a 13″ wide R&D impregnation line. A 107 cm (42″) wide carbon fabric and PMR-II-50 resin solution with 72 ± 2 %MSC were supplied to YLA by Sigmatex and Maverick Corp., respectively. The resin solution was concentrated further at YLA by vacuum distilling off methanol to predetermined levels. The hot-melt prepreg is manufactured by pouring the concentrated viscous resin between two nips rollers while the fabric and paper liners are continuously fed together as illustrated in Figure 2. The resin content on prepreg is controlled by the combination of the width of the gap between nip rollers, nip roller temperature and line speed. The process variables in the hot-melt prepregging lines were optimized based on resin viscosity, the target resin content in the prepreg, and structural quality of the prepreg after several trial runs with the PMR-II-50 resin systems (Table II).

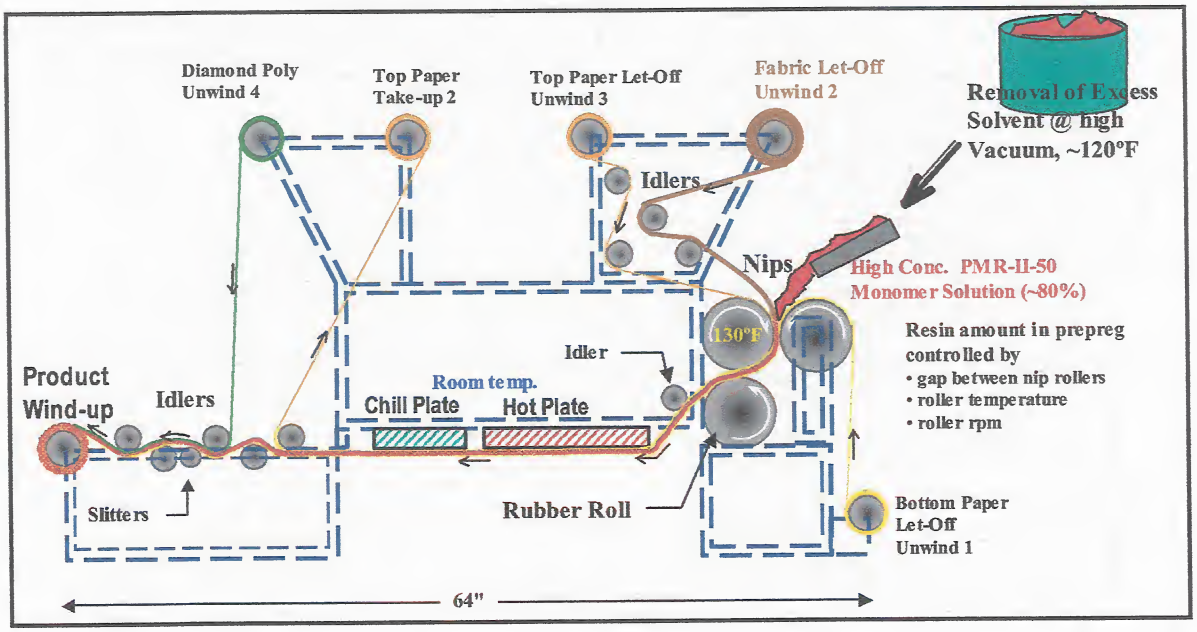

Figure 2 Schematic Diagram of the Commercial Hot-melt Prepregging Line at YLA

Table II Optimized Hot-melt Prepregging Process Variables

Run ID	Resin % MSC	Prepreg Process								Prepergger Type
		Nip Rolls			Platten		Compaction Rolls		Speed	
		Temp	Pressure	Gap	Hot	Chill	Temp	Pressure	ft/min	
HP91S	91	131 °F	low	14 mils	RT	RT	NA	NA	3.5	60" wide Commercial
HP82S	82	130 °F	100 psi	14 mils	130 °F	RT	130 °F	65 °F	3.0	13" wide R&D
HP82D	82									
HP79S	79									
HP79D	79									

As a quality control, the high pressure liquid chromatography (HPLC) analysis was performed to characterize the potential chemical aging product formation of the resin solutions at each step of

the process using a Beckman 167 System following the standard procedure described elsewhere (4). The results are summarized and compared to fresh resin in Table III. PMR-II-50 resins that were hot-melt prepregged showed more aging by-products when compared to the freshly prepared prepolymer. This is an indication that some condensation reactions have taken place during the hot-melt process. Scheiman & Alston (10-11) have reported these prepolymer 'aging' processes in earlier studies. Compared to previous HPLC analyses, the amount of aging products from the hot-melt process was insignificant (4). It was also reported that its effects on composite fabrication and quality was benign, but that was not conclusive and further details should be confirmed with more systematic investigation.

Table III HPLC Compositional Analysis Results of PMR-II-50 Resin at various Stages

	% Area (0.1% or higher only)				Normalized by HFDE, %			# of Aging Product/Adducts
	PPDA	NE	HFDE	Others	PPDA	NE	Others	
BEFORE PREPREGGING	*1. Fresh Resin; As-received, 74%MSC Newly formulated resin*							
	7.1	2.3	86.5	4.1	8.2	2.7	4.7	5
	2. 2. Solvent extracted Resin 1; 79%MSC							
	3.4	1.0	88.3	7.3	3.9	1.1	8.3	3
	3. Solvent extracted Resin 2; 82%MSC							
	5.0	0.5	87.6	6.9	5.7	0.6	7.9	5
	4. Solvent extracted Resin 3; 82%MSC							
	4.3	0.6	81.6	13.6	5.2	0.7	16.6	5
AFTER PREPREGGING	*1. Extracted resin from 82%MSC, One-pass prepreg*							
	4.9	2.0	79.0	14.2	6.2	2.5	18.0	12
	2. Extracted resin from 82%MSC, Two-pass prepreg							
	4.4	0.9	85.4	9.4	5.1	1.0	11.0	16
	3. Extracted resin from 79%MSC, One-pass prepreg							
	3.9	0.5	83.2	12.4	4.7	0.5	14.9	21
	4. Extracted resin from 79%MSC, Two-pass prepreg							
	3.9	0.7	85.1	10.3	4.5	0.8	12.1	21

2.4 Composite Curing Processes and Non-Destructive Evaluation The composite panels were fabricated using two molding processes: hot pressed in hydraulic press and autoclave. Twelve ply laminates (30.5 cm × 30.5 cm) with a cross ply configuration of $[0_f, 90_f, 90_f, 0_f, 0_f, 90_f]_{1S}$ were laid-up. This configuration was selected as the stiffness driven ply structure for this study. The lay-up was balanced, symmetric and optimized to reduce the amount of fiber crimping lines so that residual stresses that cause panel warpage would be minimized (16). For both molding processes, prepreg tapes were hand laid-up and B-staged at 204°C for 1 hr while the laminate was under a dead weight of about (30.5 cm × 30.5 cm × 1.3 cm) steel sheet in a picture-frame metal mold. B-staged laminates were vacuum-bagged using the standardized cure cycles shown in Figure 3. The hot-press molded composite panels were fabricated at GRC using a computer controlled press system. The autoclave molding was conducted at Air Force Research Lab at Dayton, OH using the same cure t-T cycles. The consolidation pressure was the primary difference between the two processes. Hot-press laminates were consolidated with 3.45 MPa (500 psi) and autoclaved panels were at 1.38 MPa (200 psi). All cured panels were then

dried in vacuum oven at 120 °C for two days and postcured at 371 °C (700 °F) in air for 16 hrs. Laminates were C-scanned using an ULTRAPAC-AD-500 from Physical Acoustics with 5MHz probe. Panels were C-scanned before and after postcure.

2.5 Test Methods Due to significant quality variation within composite panels test specimens were referenced to their panel's C-scan image. The quality of each specimen is then correlated with mechanical and physical properties. Figure 4 shows the specimen location layouts on the actual C-scan images of various composite panels (postcured).

Void content and fiber volume fraction (FVF) of composites were determined by the acid digestion method in ASTM D 3171. Three 3/4"×3/4" sections were collected from various C-scan quality regions. The glass transition temperatures, thermal degradation temperature, and dynamic mechanical properties of the cured and postcured composites were determined by standard thermal analysis techniques including dynamic mechanical analysis (TA Instrument 2980 DMA) and thermal gravimetric analysis (TA Instrument 2950 TGA HR). All composite mechanical properties were measured using an Instron test frame with Series IX Automated Materials Testing System followed by the standard test procedures listed in Figure 1. All specimens were dried at 120 °C for 24 hrs before all room temperature and 316 °C mechanical tests.

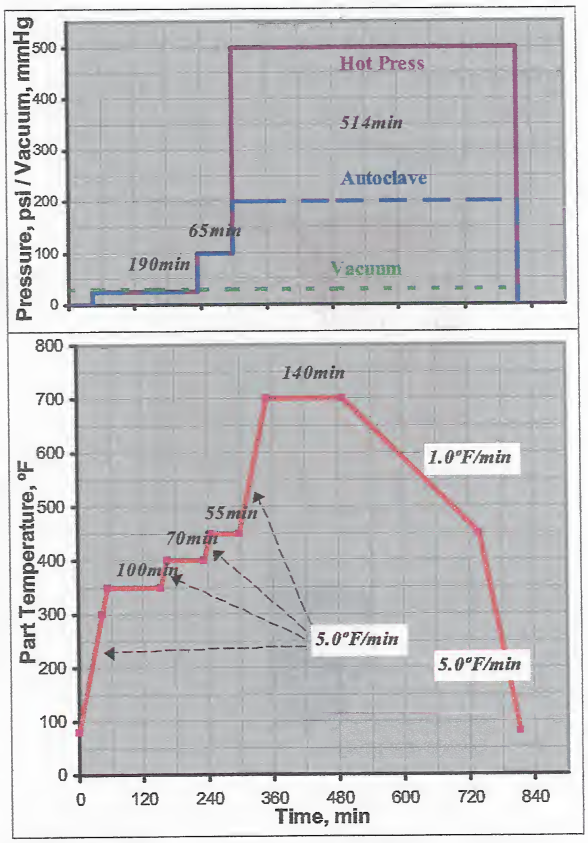

Figure 3 Composite Cure Cycles for Hot Press and Autoclave Process

3. RESULTS AND DISCUSSIONS

3.1 Composite Panel Quality The quality of the composite panels was assessed by non-destructive C-scan analysis and standard physical and thermal properties as summarized in Table IV and Figure 4. The C-scan image is color coded in terms of % transmission: 100% being the highest quality (the color images available from the CD version of SAMPE proceedings). In general, the hot-melt prepreg composites exhibit poorer C-scan quality than the solution prepreg composites. This was presumably due to poor wetting, less compaction, or ultimately higher void contents. However, the C-scan quality of the hot-melt prepreg composites was improved significantly with composites made by the prepregs with lower MSC resin solutions for both composite processing methods. This highlights the importance of the fiber-resin interfacial wetting. Comparing C-scan images for hot pressed versus autoclaved panels in Figure 4, it is

clear that the consolidation pressure is another major factor on composite quality. However, HP91S-P panel (higher viscosity resin used in prepregging) in the Figure 4 shows that the increased consolidation pressure by itself was not enough to produce a good quality composite. This also suggests that resin wetting is mostly determined at the prepregging stage when fiber surfaces were first contacted with monomer resin solution. Further, good wetting is more important than any other process conditions. PMC quality was poorest when hot-melt prepreg was autoclave molded. Note, that the double pass of the hot-melt prepreg did not significantly alter prepreg quality.

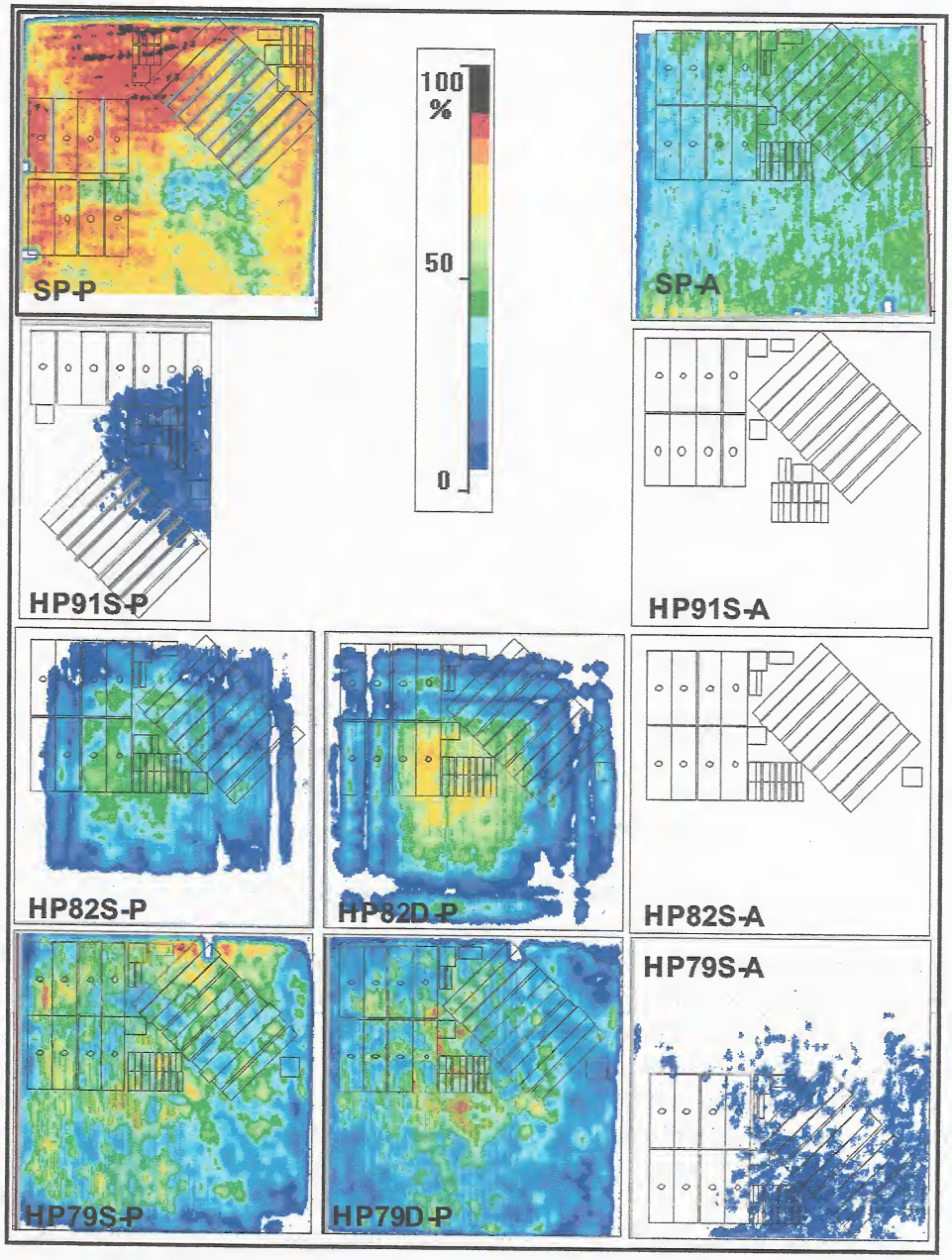

Figure 4 C-scan Images of Composite Panels from Various Prepregs

Table IV Overall Physical-Thermal Properties

Processing Code	Panel Thickness, mm	Tg, °C		Tβ, °C	TGA	Void	F.V.F, %
		G' onset	Tan δ	G" Peak	Td, °C	Content %	
SP-P	2.49 ± 0.05	374 ± 0	400 ± 0	154 ± 5	565	1.1 ± 0.7	58 ± 0.5
SP-A	2.51 ± 0.05	379 ± 3	403 ± 2	145 ± 11	559	1.5 ± 0.4	59 ± 1
HP91S-P	2.49 ± 0.05	373 ± 0	399 ± 1	148 ± 3	572	3 ± 1.6	57 ± 1
HP91S-A	2.69 ± 0.03	381 ± 0	422 ± 1	158 ± 1	579	7.8 ± 2.1	57 ± 1.4
HP82S-P	2.87 ± 0.15	371 ± 0	397 ± 1	141 ± 6	569	2.1 ± 0.6	54 ± 0.6
HP82D-P	3.14 ± 0.15	370 ± 1	394 ± 1	140 ± 4	568	1.4 ± 0.2	48 ± 1.2
HP82S-A	2.95 ± 0.15	382 ± 1	407 ± 0	150 ± 0	567	8.2 ± 2.2	55 ± 1.2
HP79S-P	2.39 ± 0.08	377 ± 1	404 ± 1	159 ± 6	568	2 ± 0.6	63 ± 0.1
HP79D-P	2.44 ± 0.08	378 ± 4	404 ± 1	151 ± 1	566	1.7 ± 0.3	62 ± 0.7
HP79S-A	2.59 ± 0.12	382 ± 1	404 ± 1	156 ± 2	567	3.8 ± 0.3	62 ± 0.8

The average void content of each composite type is listed in Table IV. Note that most standard deviations are quite high with a coefficient of variation (COV) of 20-60%. This variation was due to sampling differences. The three acid digestion samples were collected from a wide range of composite quality within each panel as shown in Figure 4. For more meaningful analysis, the void contents were plotted versus C-scan % transmission as shown in Figure 5. From previous studies (3-4), the correlation between C-scan % transmission and void content was primarily dependent on panel thickness and fiber type. For the composite systems studied, all data points exhibited the same trend as seen in the plot in Fig.5 regardless of prepreg type or molding condition. However, they are separated into two distinctive groups. Void contents of most hot pressed composites lined up approximately at 2% or lower level while autoclaved composites had higher void contents. The exception was one of the composites made from the solution prepreg. Certainly, higher consolidation or processing pressure lowered the composite void content, possibly by forcibly removing trapped voids or improving fiber wetting. At lower consolidation pressures used in autoclave molding, the void content was reduced consistently when prepregs were produced from lower MSC wt%. Once again, this suggests that wetting at prepregging stage is more important than any altering other curing conditions. Another observation seen in Figure 5 is that in the lower void content group, C-scan % transmissions of all composite types varied by more than 70% even though void content changed by only 0.5-1.0 %. In the higher void content group, however, C-scan % transmission only showed a small change, about 10%, while void content varied by more than 8%. This simply

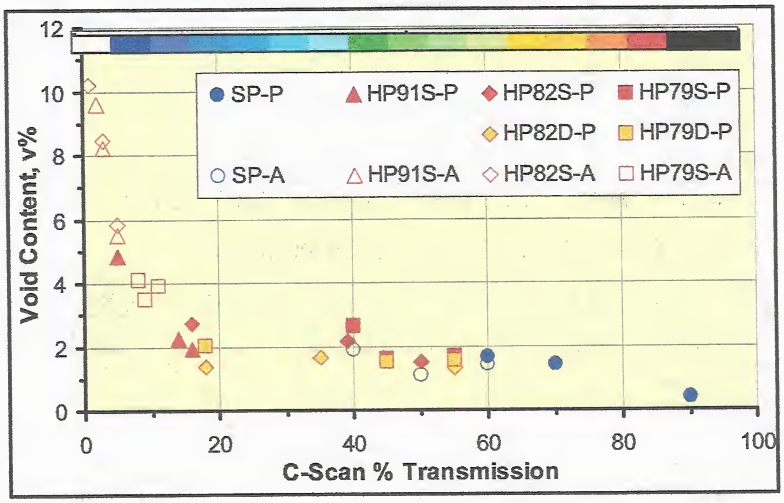

Figure 5 Void Content vs. C-scan % Transmission for Various Composites

suggests that C-scan was more sensitive to general composite quality when composite void content was low.

The physical and thermal properties are summarized in Table IV. Variations in panel thickness were due to changes in consolidation pressure and resin content. Comparing similar ply configurations of the same fiber-resin composite, the panel thickness can be used as another indicator of panel quality. Autoclave-cured panels were slightly thicker than the hot pressed laminates. Composite thermal properties, such as glass transition, T_g, secondary glass transition, T_β, and thermal degradation, Td, were similar for both hot-melt and solution prepreg and for autoclave and hot press cured laminates.

3.2 Composite Properties Composite mechanical properties measured at room temperature and at 316 °C are summarized in Tables V and VI, respectively. Figures 6 and 7 show overall trends of various composite properties against processing options by a percent change from normalizing each value with properties of baseline composite, SP-P (hot pressed composite with solution prepreg), for room temperature and 316 °C properties, respectively. Comparison of composite mechanical properties shows the similar trends as the composite quality analyses.

Table V Overall RT Mechanical Properties

Processing Code	In-plane Shear Strength, psi	OHC Properties, ksi		SBS Strength, psi	DMA Storage Modulus, G', ksi
		Ini. Modulus	Strength		
SP-P	9436 ± 136	1613 ± 28	28 ± 1.9	5947 ± 188	3719 ± 892
SP-A	n/a ±	n/a ±	n/a ±	n/a ±	2073 ± 267
HP91S-P	9583 ± 250	1442 ± 47	22.5 ± 2	5984 ± 186	2682 ± 164
HP91S-A	4343 ± 78	259 ± 128	6.8 ± 0.7	n/a*	1711 ± 410
HP82S-P	9693 ± 107	1410 ± 85	25.6 ± 1.5	6159 ± 245	2603 ± 420
HP82D-P	12500 ± 2096	1268 ± 21	26.9 ± 1.7	6073 ± 154	1812 ± 21
HP82S-A	7215 ± 471	1087 ± 106	14.3 ± 2.9	4336 ± 272	1704 ± 236
HP79S-P	10996 ± 649	1572 ± 55	23.6 ± 0.9	5261 ± 282	2305 ± 656
HP79D-P	11198 ± 401	1679 ± 37	28.4 ± 1.3	5266 ± 310	2392 ± 185
HP79S-A	9225 ± 1222	1429 ± 49	18.5 ± 1.8	3933 ± 511	2719 ± 892

* samples failed by compresion mode w/ plastic deformation

Table VI Overall 316°C Mechanical Properties

Processing Code	In-plane Shear Strength, psi	OHC Properties, ksi		SBS Strength, psi	DMA Storage Modulus, G', ksi
		Ini. Modulus	Strength		
SP-P	7777 ± 128	1510 ± 143	21.8 ± 1.7	4862 ± 148	3255 ± 789
SP-A	n/a ±	n/a ±	n/a ±	n/a ±	1745 ± 254
HP91S-P	8434 ± 340	1267 ± 144	15.8 ± 2.3	4349 ± 86	2211 ± 113
HP91S-A	3537 ± 133	241 ± 299	5.4 ± 0.1	n/a*	1457 ± 297
HP82S-P	8202 ± 430	1364 ± 55	19.4 ± 1.4	4893 ± 100	2175 ± 349
HP82D-P	11355 ± 811	1168 ± 69	19.4 ± 0.8	4697 ± 90	1522 ± 41
HP82S-A	5707 ± 260	784 ± 410	10.9 ± 1.9	3165 ± 126	1370 ± 154
HP79S-P	8322 ± 512	1458 ± 124	19.7 ± 0.8	4190 ± 150	2059 ± 451
HP79D-P	8785 ± 230	1575 ± 73	21 ± 1.2	4485 ± 210	2073 ± 123
HP79S-A	7010 ± 1084	1303 ± 130	13.9 ± 0.9	2941 ± 302	2240 ± 666

* samples failed by compresion mode w/ plastic deformation

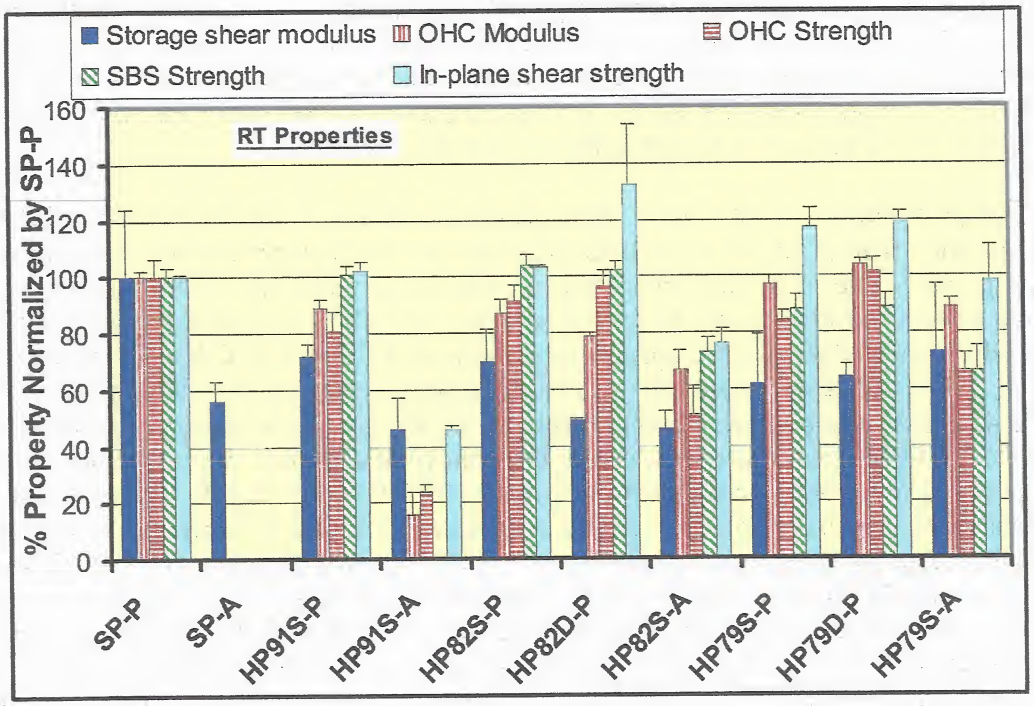

Figure 6 Overall RT Mechanical Properties Comparison

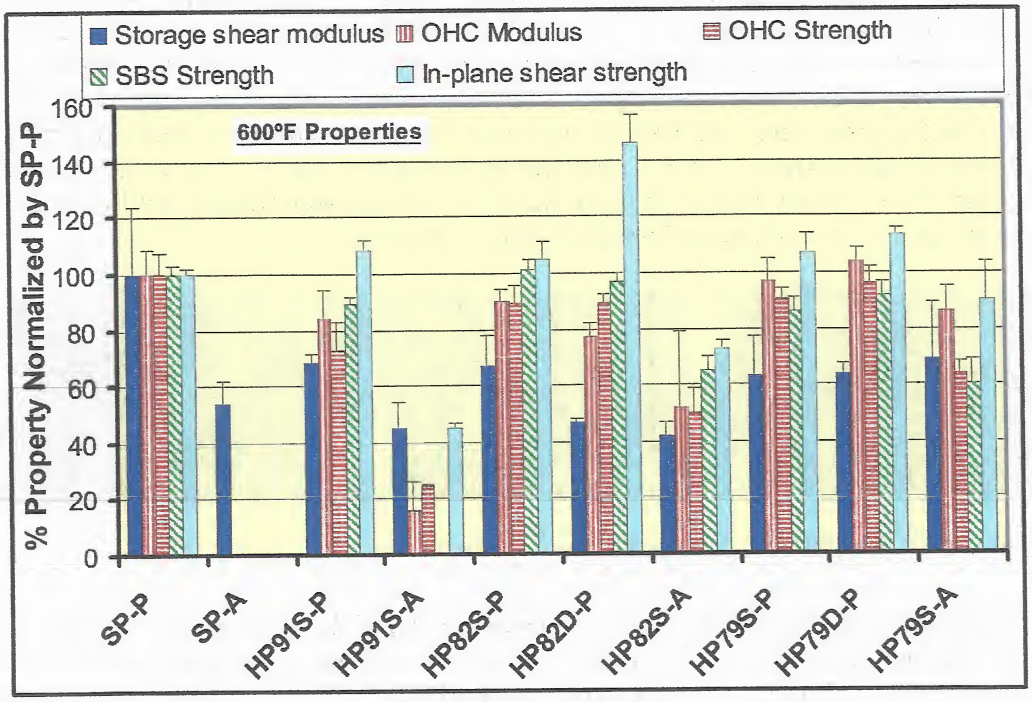

Figure 7 Overall 316°C Mechanical Properties Comparison

In general, composites from hot melt prepregs had lower mechanical strengths and stiffness than composites fabricated from solution prepreg. The laminates produced from hot-melt prepreg and processed with an autoclave had the poorest properties. Lowering the resin solution viscosity for the hot-melt prepregs, especially for the autoclaved panels, made significant improvements for both RT and 316 °C properties as seen in Figures 6 and 7.

DMA storage moduli at RT and 316 °C were obtained from single cantilever mode with 1Hz and oscillation amplitude of 20 μm. Open hole compression (OHC) properties were considered as an off-axis property due to triaxial stress field developed at notch tip. These properties were sensitive to both the fiber-matrix interfacial qualities and fiber alignment. Figure 8 shows the typical edge views of tested OHC specimens (from both RT and 316 °C test) at the center hole location. The baseline composite, SP-P failed by a typical shear mode with a clear kink band close to 45° of the compressive loading direction, i.e., the composite compressive strength was controlled by fiber shear strength, which is the typical OHC failure mode of fiber reinforced composites. The hot-melt, autoclaved composites, HP91S-A had widely scattered and wider kink bands than hot press, solution prepregged panels. This was probably due to fiber-matrix debonding caused by lower interfacial strength. All other composites, such as HP82S-P failed by a mixed mode of kink bands and localized interlaminar splitting. This is indicative of lower interfacial strength. Both mixed modes formed more extended kink bands with smaller angle toward the loading axis. Further, this mode change was consistent with property changes. The ± 45° off-axis tension test measures in-plane shear strength, but not just by matrix or interface properties alone. Fiber netting formation from the woven fabric structure also contributes to the in-plane shear properties. The strength is also influenced by fiber orientation. All the composites tested formed the netting. The abnormally high in-plane strength of HP82D-P might be due to fiber misalignment, but the high in-plane strength of several hot-melt prepreg composites can be attributed to more intensified netting resulted from hot-melt processes where the impregnated fabric passed through various compaction rolls, especially in double pass prepregs. On the other hand, the SBS strength was the most matrix dominant property and the strength was closely correlated with C-scan quality (Figures 9 and 10). Interestingly, composite interlaminar shear strength (ILSS) does not begin to decrease significantly until composite void volumes are greater than 4% regardless of the type of prepreg.

Figure 8 Digital Micrographs Showing OHC Failure Modes

3.2.1 Solution vs. Hot-melt and Effects of Monomer Solid Concentration Clearly, solution prepregs provided better composite quality. However, composite properties, such as in-plane shear strength and ILSS, hot-melt prepreg performed equal to or better than solution prepreg. Since prepreg quality was determined by the diffusion-controlled wetting, improving the hot-melt prepregging process can be further optimized to improve the wetting. In fact, composite

quality and properties improved significantly by lowering the monomer concentration from 91% to 79%.

3.2.2 Effects of Double Pass on Hot-melt Prepreg Slight improvements in composite quality and properties resulted from passing the prepreg through compaction rollers twice despite the 3-4% loss in solvent (methanol) content from the first run. An increase in the in-plane shear strength for composites tested at RT and 316 °C data by double-pass prepregging suggests that the additional compaction enhances intralaminar adhesion. This additional compaction also lowered void content as shown in Table IV.

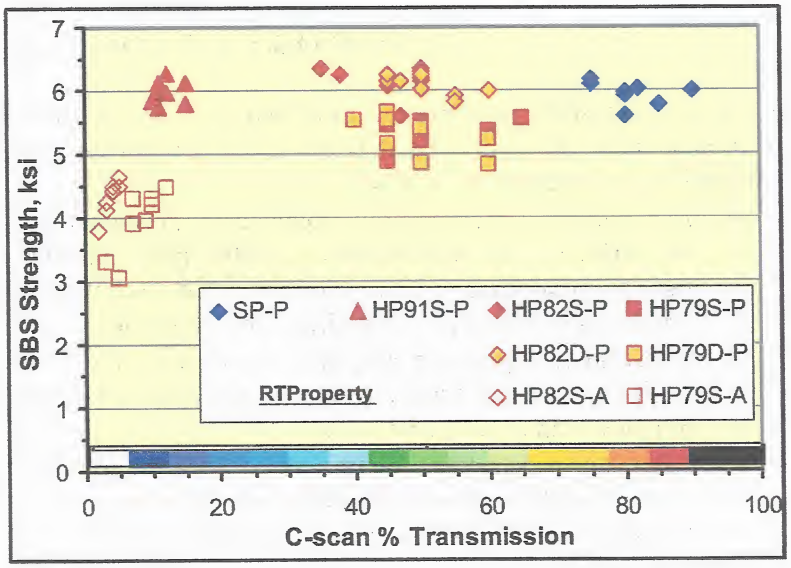

Figure 9 RT Interlaminar Shear Strength vs. C-scan % Transmission

3.2.3 Effects of Composite Cure Method The most important factor that improved composite quality and properties resulted from the change in consolidation pressure during processing. Wherein an increase in pressure from 1.38 MPa (200 psi) to 3.45 MPa (500 psi) improved both composite characteristics. This was particularly noticeable for hot melt prepreg. Composites fabricated with the higher % MSC hot-melt prepregs (HP91 and HP82) using an autoclave with 1.38 MPa consolidation pressure had higher void content, lower stiffness, and strengths and were not acceptable. Whereas, composites made from the lower % MSC (79%) prepreg (HP79-A) had properties that were closer to the control composites. Therefore, thorough wetting and high compaction are both required for producing high quality composite panels from these types of polyimides and graphite fiber fabrics of similar fiber areal weight. Moreover,

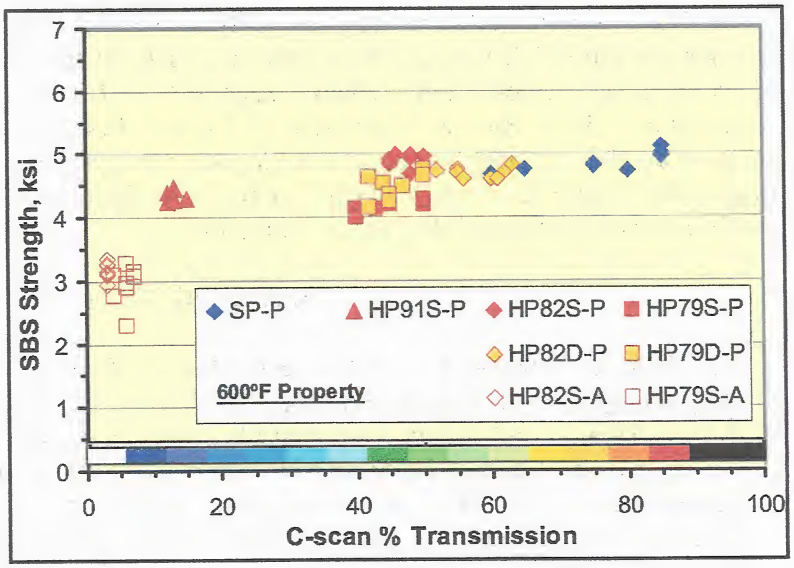

Figure 10 316°C Interlaminar Shear Strength vs. C-scan % Transmission

prepregs that were wet effectively, as in solution prepared prepreg, are not as significantly affected by decreases in processing pressure.

4. SUMMARY AND CONCLUSIONS

The composite quality and properties of two commercial prepregging methods (solution and hot-melt processes) for M40J/PMR-II-50 fabric composites were evaluated and the important conclusions are summarized below:

- In general, hot-melt prepreg produced composites of poorer quality than solution prepreg. The primary reason for this quality difference was due to poor fiber wetting in the hot-melt prepreg process. Thus, lowering resin viscosity in hot-melt prepregging significantly improved both composite qualities and properties.
- Consolidation pressure played a significant role in producing good quality composite. Both the extent of fiber wetting and the optimum consolidation pressure are necessary to produce high quality composites.
- After optimizing fiber wetting and composite processing pressures, the composite void contents and mechanical strengths were similar.
- Finally, composite ILSS suffers when the void content is greater than ~4% regardless of prepreg type.

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